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PATENT SPECIFICATION

Convention Date (Germany): Feb. 16, 1929.

355,210

Application Date (in United Kingdom): Feb. 17, 1930. No. 5844 / 30.

Complete not Accepted.



COMPLETE SPECIFICATION.

**Processes for Recovering Higher Hydrocarbons and Hydrogen or
Gases Containing Hydrogen.**

We, RUHRCHEMIE AKTIENGESELLSCHAFT, of Oberhausen-Holten, Germany, a Body Corporate organised and existing under the Laws of the German State, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:

The need frequently arises for extracting the hydrogen content in gases containing hydrocarbons in chemical combination as free hydrogen whether in order to employ the hydrogen for chemical conversions for example for the synthesis of ammonia or whether it be in order to correct the thermal value or the combustion properties of the gases containing hydrocarbons by enrichment with molecular hydrogen.

It has been found that gases which contain hydrocarbons such as methane, ethylene, acetylene or their homologues (such for example as coke oven gas, natural gas, the waste gas from benzol synthesis or the methane bearing gases resulting from the catalytic conversion of mixtures of carbon monoxide and hydrogen can be converted into gases having a high hydrogen content by exposing these gases to the action of electrical fields (such as high frequency oscillations) at a pressure depending upon the nature of the fields. Under this treatment the said hydrocarbons part with hydrogen, while at the same time polymerisation products possibly of a recoverable nature, are formed. Thus the extremely uneconomical loss of calorific power which is unavoidable in the otherwise usual cracking process is avoided for example by separating out inferior solid carbon.

When treating methane or methane-containing gases by heat in the use of electrical fields, a gas under 40 mm.

pressure is by way of example, subjected to a high voltage alternating current discharge, alternating current discharges for example of 7,000 volts being employed. In this connection about 20% of the methane is converted into acetylene, tar, and products similar to benzol hydrocarbons. At the same time hydrogen is produced to the extent of 25% by volume. By washing with cooled acetone, the acetylene and the other products of conversion up to the hydrogen can be removed to 100%. The hydrogen thus obtained may be at once employed for technical purposes.

It is known to convert gases containing hydrocarbons, and which contain methane, ethylene, acetylene or their homologues, if necessary in mutual admixture, such as coke oven gas, rich gas from coke gas decomposing apparatus, natural gas, exhaust gas from benzene synthesis, inter alia by a short heating to temperatures over 900° without the use of catalysts, into gases which are rich in hydrogen and contain a certain content of extractable tarry and benzol hydrocarbons (see Fischer, "Synthese von Benzolkohlenwasserstoffen"—Synthesis of benzol hydrocarbons—"Brennstoffchemie", 1.10.1928). It is also known that the said gases containing hydrocarbons can be transformed into polymerisation products such as acetylene, hydrocarbons, benzol hydrocarbons and tar hydrocarbons, hydrogen being split off by the action of electrical fields (for example in the form of dull emission discharges) (see "Technische Rundschau der Bergwerkszeitung" of 3.2.1929).

According to the invention the splitting off of hydrogen with formation of recoverable polymerisation products proceeds in a particularly smooth manner and with good yield if the electric treatment be combined with the heat treatment

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in such a way that the heating is effected simultaneously with the action of the electrical fields, or directly thereafter. By means of this combined process, the yield of recoverable liquid and solid hydrocarbons can be substantially increased in comparison with the yield obtainable by applying the processes singly, with the same expenditure of heat or electrical energy.

An example of conversion by the joint use of heat and electricity may be mentioned in which it is desired to produce as the final product a gas having a maximum content of hydrogen. A gas for example of a composition of 0.1% of C_2H_m , 0.7% of O_2 , 1.7% of CO , 90% of CH_4 and 7.5% of N_2 is subjected to the process. If this gas be subjected at a temperature of 450° to the action of an alternating current for example of 8,000 volts and 180 millamps at a pressure of 20 mm. mercury, a gas is obtained which, after the removal of the undesirable components in known manner has the following composition: 0.2% of C_2H_m , 1.3% of CO , 90% of H_2 , 4% of CH_4 and 4.5% of N_2 .

If the content of hydrogen is to be smaller, the intensity of the effect or its duration must be reduced accordingly. Thus for instance under otherwise similar conditions a reduction in the electrical high voltage field to 4,000 volts produces only about a 60% conversion of the methane, while the conversion when using the 8,000 volt field is almost 100%. A similar increase in temperature has the effect when the field strengths are low of increasing the splitting up to an important degree. At high temperatures such as about 800° and field strength which even at low temperatures produce practically 100% conversion of the methane, the products of conversion that are separated out change. At high temperatures more liquid polymerisates are obtained, until when exceeding a certain temperature or duration of treatment which are dependent upon each other, solid carbon is precipitated.

The gases enriched with hydrogen may be further treated according to the purpose for which they are intended.

For example the hydrogen can be freed by supercooling or other known processes of similar character, from undesirable impurities (decomposition products) which may if desired be economically utilised and rendered suitable for catalytic operations. A gas is for example treated which has been obtained by the combined heat and electrical treatment and which has a hydrogen content of 90%. By corre-

spondingly conducting the cooling according to the Concordia-Linde-Bronn process, while adding the required nitrogen, a gas is produced from the gas mixture containing 90% hydrogen, 70% which gas contains 25% nitrogen and 75% hydrogen. As the removal of the remaining constituents succeeds to such extent that carbon monoxide, methane and the like are present only to the extent of from 0.01% to 0.05%, the mixture of nitrogen and hydrogen obtained may be immediately employed for the synthesis of ammonia.

The process may also be employed for transforming (for example) gas high in hydrocarbons resulting from the decomposition of coke gas, or from a source of natural gas with simultaneous recovery of economically utilisable condensation products, in certain cases into a gas which closely approximates to coal gas in composition and applicability and is suitable for gas-supply purposes.

Another object attainable by the proposed treatment may consist in applying the said process to the control of the calorific value of gas from a distant source of supply, in that a fraction enriched with hydrocarbons is obtained from the whole or a portion of said gas, or from a gas of similar composition, in a gas decomposing plant operating according to a known process, and that said fraction is exposed to the action of the electrical fields. After conversion the undesired constituents in the resulting gas are removed by known methods, and the gas either by itself or in admixture with the decomposition fractions which are low in hydrocarbons, is added to the gas from the distant source, the calorific value of which can be thus modified to the desired extent according to the amount of the gas drawn off for treatment and the degree to which the decomposition has been carried. Finally a rich gas high in hydrocarbons and difficult to utilise can be completely transformed into hydrogen and condensation products by repeatedly subjecting it to the electrical treatment after a previous purification in each case.

A feature of high technical importance is that in the electrical treatment of the gases, the composition of the gas finally obtained can be adapted in a variety of ways to the purpose in view, by regulating the pressure, intensification, duration and method of action of the electrical fields. Gaseous constituents which are undesirable in relation with the purpose in view may be eliminated from the decomposed gas by subsequently applied known methods of purification.

- It should also be noted that not only the hydrogen produced, from hydrocarbons or mixtures containing the same by the action of electrical fields can be employed, after suitable preliminary treatment for catalytic or other purposes, but that hydrogen obtained by splitting hydrocarbons in any other way can be similarly employed.
- 10** Having now particularly described and ascertained the nature of our said invention and in what manner the same is to be performed, we declare that what we claim is:—
- 15** 1. A method of carrying out the specified process with special application in the recovery of hydrogen or gases high in hydrogen, characterised in that the gases are subjected under a definitely controlled pressure to the action of electromagnetic fields (such as high-frequency oscillations), and that the reaction products unsuitable for the purpose in view are eliminated from the converted gas, and may if desired be economically recovered in known manner.
2. A method of carrying out the specified process, characterised in that, in the electrical treatment the composition of the final gas is modified according to the final purpose in view, by regulating the pressure and the duration, intensity and nature of the operation of the electrical fields.
- 35 3. A method of carrying out the specified process, characterised in that the gases are subjected simultaneously or in succession to the action of an electrical treatment and a heat treatment, in which according to the nature of the gases brought into reaction and the nature of the desired final products, the duration of the treatment, the pressure, strength and nature of the electromagnetic fields, and also the duration and temperature applied in the heat treatment are regulated.
4. Process according to the preceding claims, characterised in that the hydrogen produced during conversion is rendered of use for catalytic conversions after the removal of the remainder of the undesirable gaseous constituents by low temperature cooling or similar processes.
5. Process according to the preceding claims, characterised in that the gases produced during conversion are applied to gas supply.
6. Process according to the preceding claims, characterised in that gases containing hydrocarbons which are drawn off for example as a partial stream from a distant gas supply or are obtained in aliquot quantity and similar composition from a different gas source, are subjected to the described conversion process, if necessary after first being enriched with hydrocarbons, in a gas decomposing plant operating by a known process, until by admixing the conversion gases from which undesirable admixtures have been removed with the gas from the distant supply the calorific value is adjusted to a determined point.
7. Process according to the preceding claims, characterised in that the available gases which contain hydrocarbons, if necessary after first being purified of the undesirable reaction products are repeatedly subjected to the conversion process.
8. Combined process for the recovery of higher hydrocarbons from gases which contain methane, ethylene, acetylene or their homologues if necessary in mutual admixture, characterised in that the gases are exposed simultaneously to a short period treatment at temperatures above 900° and to the action of electromagnetic fields (for example dull emission discharges or high frequency oscillations).
9. Process according to claim 1, characterised in that the action of electromagnetic fields take place first, followed by the heat treatment.

Dated this 17th day of February, 1930.

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